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Gamma-ray Measurements of a 6-kg Neptunium Sphere

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Introduction

In order to better determine the properties of ^{237}Np for criticality safety and nuclear nonproliferation, especially its critical mass, 6070-gram solid sphere was cast on 15 May 2001 in a hot cell [1]. The casting sprue was cut off on a lathe and the casting ground to a final diameter of 8.29 cm. The sphere was enclosed in a spherical tungsten shell 0.523-cm thick to reduce the gamma-ray dose. The neptunium and the tungsten were doubly encapsulated in welded, spherical nickel shells, each 0.381-cm thick. The sprue material was analyzed by mass spectrometry. Here we report the results of the first gamma-ray measurements of this unique item.

Measurements

We measured the gamma-ray output on four different dates in 2001, 30 May, 29 June, 27 July, and 25 September, to observe the expected growth of ^{233}Pa and other daughter isotopes. We used two p-type germanium detectors, a nominal 80% detector set up at distances in the range 380–410 cm from the center of the neptunium sphere and a nominal 140% detector at a distance of 141 cm with a 0.74-cm-thick machinable tungsten filter. The larger detector was set up to measure any high-energy (> 3 MeV) gamma rays that might be present due to neutron capture and charged particle reactions, but none were observed. Measurement live times were three to four hours.

The primary isotopes found in the sphere were ^{237}Np ($t_{1/2}=2.1 \times 10^6$ years) and its daughter ^{233}Pa ($t_{1/2}=27$ days) and the contaminant ^{243}Am ($t_{1/2}=7370$ years) and its daughter ^{239}Np ($t_{1/2}=2.4$ days). Other isotopes found include $^{233,238}\text{U}$, $^{239,240}\text{Pu}$, and ^{241}Am . It is highly likely that ^{235}U is also present, but the relatively weak 186-keV gamma ray from ^{235}U is overwhelmed by the intense gamma rays from ^{233}Pa and ^{239}Np .

Table 1 shows the absolute emission rate of three of the primary gamma rays from ^{233}Pa and ^{239}Np . The ^{233}Pa emission rates are essentially constant in time. Because the spatial distribution is not uniform (discussed below), we believe that the variations are due to slight angular offsets from one measurement to the next, exposing more or less of the sprue area.

Table 1. Emission rates (γ/s) as a function of time for the two primary isotopes

| Date | ^{239}Np , 277 keV | ^{233}Pa , 300 keV | ^{233}Pa , 398 keV |
|--------------|-----------------------------|-----------------------------|-----------------------------|
| 30 May 2001 | 3.07×10^6 | 2.03×10^7 | 1.59×10^7 |
| 29 June 2001 | 3.04×10^6 | 2.08×10^7 | 1.64×10^7 |
| 27 July 2001 | $3.xx \times 10^6$ | $2.xx \times 10^7$ | $1.xx \times 10^7$ |
| 25 Sept 2001 | 3.87×10^6 | 1.95×10^7 | 1.58×10^7 |

We also attempted to measure the isotopic composition of the plutonium in the sphere using the gamma rays in the 640-keV region, the only region where we could observe

gamma rays from plutonium. We obtained 0.10 ± 0.08 for the ratio of ^{240}Pu to ^{239}Pu . We also measured 0.014 ± 0.004 for the ratio of ^{241}Am to ^{239}Pu . These values are consistent with the mass spectrometer values for the sprue material of 0.071 and 0.017, respectively. Combining the plutonium results with the measured neutron output of 1.2×10^4 neutrons/s and measured multiplication of 1.9 [2], we estimate that the sphere contains 63 grams of plutonium, a value much higher than the 2.1 grams indicated by the mass spectroscopy of the sprue material.

The behavior of the five elements found by gamma-ray spectroscopy (Pa, U, Np, Pu, and Am) should have been quite different while the neptunium was in the molten state. Only plutonium has a melting point (640°C) lower than neptunium (641°C); the other three melt at substantially higher temperatures, 1100°C or above. Therefore, because the liquid density of the neptunium was estimated to be $\rho \sim 17\text{--}18\text{ g/cc}$, the uranium ($\rho = 18.7\text{ g/cc}$) should have tended to sink toward the bottom of the sphere. On the other hand, the americium ($\rho = 13.7\text{ g/cc}$) and the protactinium ($\rho = 15.4\text{ g/cc}$) should have floated toward the top, and the plutonium ($\rho_{\text{liquid}} = 16.6\text{ g/cc}$) should have stayed approximately uniformly distributed.

We measured the gamma-ray emission output at 18 points on the surface of the sphere using a 1.27-cm-diameter hole in a 5.1-cm-thick lead brick as a collimator. The spatial distribution of the ^{233}Pa was constant within the measurement errors ($\sim \pm 3\%$), which is surprising in view of its lower density. In contrast, the distribution of ^{239}Np , the short-lived daughter of ^{243}Am , was highly asymmetric, concentrated near the sprue area. The ^{243}Am concentration varied from 1680 ppm to 208 ppm. For comparison, the mass spectrometer value for the sprue was 1820 ppm.

Conclusions

The bulk of the sphere does not have the same composition as samples taken from the sprue for destructive analysis. In general, the bulk of the sphere has lower americium content but higher plutonium content than is indicated from the sprue samples. Our investigation of this unique sample will facilitate other studies, in particular, criticality studies.

Acknowledgements

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References

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